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Qualitative Light-Scattering Angular Correlations of Conglomerate Particles

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Abstract

The scattering phase functions of micrometer-size glycerol droplets containing spherical latex inclusions undergo random fluctuations with time. We measure scattering intensities in the near-forward and near-backward scattering directions and find them to have strong positive correlations during some time periods and strong negative correlations during other time periods. The characteristic time constants of these correlations are on the order of seconds. We calculate scattering correlations from two types of scattering systems. Correlations from an aggregate of two isolated spheres are generally positive, whereas correlations from a sphere containing a single spherical inclusion may be both positive and negative. Calculations of correlations from our experimental data are consistent with diffusion of inclusions within the host droplet, rather than interference effects between the inclusions.

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1. Introduction

Optical properties of levitated droplets have been studied extensively during the last 15 years; particular attention has been focused on scattering resonances in droplets [1–11], now commonly called morphology-dependent resonances. Recently, several papers have examined both elastic [12–14] and inelastic [15–21] scattering of a host droplet containing small inclusions. Unlike homogeneous droplets, a droplet containing inclusions displays a fluctuation in the scattered intensity. The nature of such fluctuations has been attributed to inclusions passing successively through regions of strong electric fields (hot spots) and weak electric fields within the host as the droplet rotates [13] and to interference of waves scattered by the inclusions within the host droplet [14,21]. Previous researchers [14,21] used the latter attributes of the scatter in photon-correlation analyses to try to determine physical characteristics of the inclusions. However, the results of these analyses were not conclusive.

In our study, we perform photon-correlation analyses, but the nature of the correlation is fundamentally different from that in previous work. Whereas previous correlation analyses on scattering data [14,21] used the autocorrelation function, we consider data taken at two different scattering angles as a function of time, and we examine the resulting correlations between the data sets. For this, we use a standard correlation function defined as

$$\rho_{xy} = \frac{\sum\limits_{i} (x_i - \bar{x})(y_i - \bar{y})}{\sqrt{\sum\limits_{i} (x_i - \bar{x})^2 \sum\limits_{i} (y_i - \bar{y})^2}}, \tag{1}$$

where x_i and y_i are the intensity signals measured at time t_i at two different scattering angles, and $\bar{x}$ and $\bar{y}$ are the time averages of these signals. The correlation function varies between $+1$ (the limit for perfectly correlated signals) and -1 (the limit for perfectly anticorrelated signals).

We try to develop some insight into the scatter by comparing the correlations that we see in the experimental scatter with those calculated with some simple theoretical models. For instance, equations describing the scattering

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of electromagnetic radiation by systems composed of individual spheres in close proximity have been derived and exploited in a number of applications [22–39], and we used these equations to model the interference of waves scattered by inclusions within a host. Parallel research has addressed the scattering from a subsystem completely enclosed by a spherical host [40–48]. By comparing the experimental correlations to those calculated with these models, we hope to gain an understanding of the scattering processes. The ultimate goal is to eventually use this technique to quantify the scattering system.

2. Experiment

Figure 1 shows a diagram of the experimental apparatus. A charged glycerol droplet with radius of $r_h \sim 10$ to 12 μm is captured in an electrodynamic trap [49]. (We generated the droplet with a spray atomizer, prepared to contain a suspension of spherical, uniform latex inclusions having a known concentration.) Levitated droplets are illuminated with a moderately focused (beam waist $\sim$1 mm) krypton-argon laser beam ($\lambda = 647.1$ nm), while simultaneously photomultiplier tubes (PMTs) detect the scattered light. The PMTs, placed in the forward-scatter ($\sim$7°) and backward-scatter ($\sim$179.5°) directions, collect light over a relatively small conical solid angle (approximately 0.1° half-angle). The detected signals are amplified, fed through a low-pass (250-Hz) filter, and digitized with an 820-Hz analog-to-digital recorder.

The relative background noise levels have been measured for all the data sets and are typically less than 0.5 percent of the droplet signals. The shot noise is the dominant noise and is typically a small percentage of the signal.
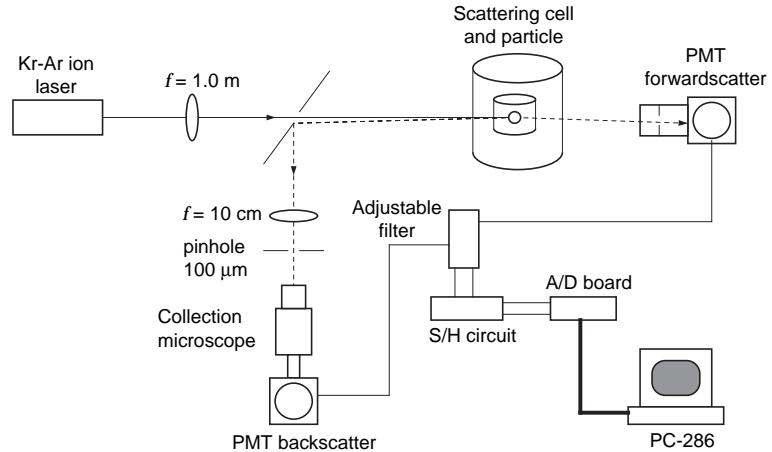
Figure 1. Schematic of experimental setup used to measure elastic scattering from micrometer-size glycerol droplets seeded with latex particles. Droplets are caught in an electrodynamic trap and are illuminated with a krypton-argon laser ($\lambda = 647.1$ nm). Scattered light is simultaneously detected by photomultiplier tubes (PMTs) placed in the forward-scatter ($\sim$7°) and backward-scatter ($\sim$179.5°) directions. These signals are amplified, digitized, and stored.

To minimize evaporation, we hold the droplets for approximately 1 hour in the electrodynamic trap so that equilibrium with the vapor within the containment vessel enclosing the trap can be attained (surfaces of the trap are coated with a sufficient amount of glycerol during the droplet-trapping process to saturate the vapor within the vessel). However, since the containment vessel is not completely air-tight (it contains holes to allow for entrance and exit of the laser beam), some vapor does escape, and the host droplet slowly evaporates at a rate of approximately 0.2 nm over a 6-s data run.

Figure 2a shows a sample of the experimental data for a droplet containing inclusions: a 2-s data set for a glycerol host ($r_h \sim 12$ μm, $n_h = 1.471$) containing approximately 170 latex inclusions ($r_i = 0.50$ μm, $n_i = 1.59$). We determine the host radius to within 5 percent by examining the distance between the diffraction rings [50]. We know the number of inclusions from the original concentration of latex in the glycerol-latex suspension. As observed previously [12–14], when the droplets are seeded with latex spheres, the signals fluctuate greatly (>50 percent) over small time scales (<1 s). One feature evident in the data is a low-frequency ($\sim$1 Hz) correlation between the backward-scatter and forward-scatter signal intensities. This correlation can be seen more clearly after the data are filtered with a 30-Hz median filter. In the data set in figure 2, the correlation is strongly positive through much of the first second, and strongly negative through much of the second second.

The data in figure 2a are fairly typical of most data sets we have recorded. There are time periods when the forward-scatter and backward-scatter signals exhibit high positive correlation, and other periods of high negative correlation. Figure 2b shows a running correlation of the data shown in figure 2a, calculated with a 0.5-s data window, after both signals are filtered with a 30-Hz median filter. The correlation varies between −0.8 and 0.7, and is typical of other correlations observed.
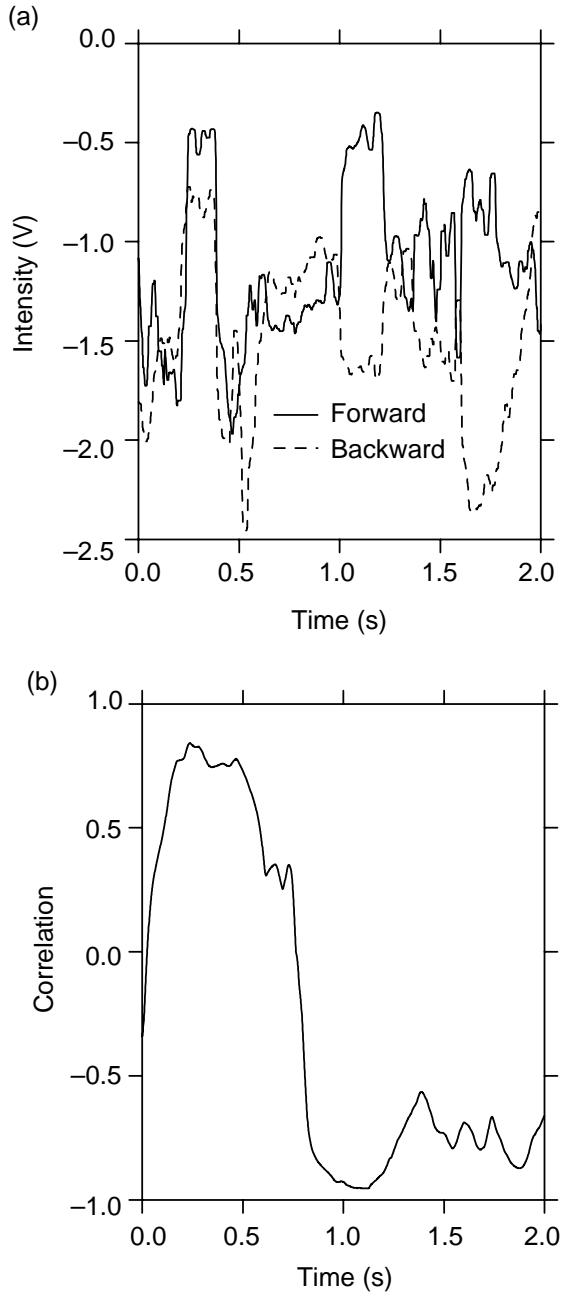
Figure 2. Experimental data showing (a) scattered intensity of an $r_h \approx 12$ μm glycerol host containing approximately 170 latex inclusions, $r_i = 0.50$ μm, in forward-scatter and backward-scatter directions as a function of time, and (b) correlation function of scattered signals.

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3.   Analysis

In order to understand the nature of the correlation of the forward and backward scatter, we use some theoretical models to calculate scattering correlations from systems having characteristics similar to our experimental system. We examine two systems: a pair of isolated spheres, and a sphere containing a nonconcentric spherical inclusion.

It has been hypothesized that the oscillations are due to interference among scattered waves from individual spheres within the host droplet [14,21]. To explore this hypothesis, we approximate the interference among individual spheres by examining the scatter from a two-sphere system [22–39], whose correlation is due solely to interference between the two spheres. Our motivation is to qualitatively explain the correlation of the considerably more complicated experimental scattering system, where there are typically between 2 and 200 interacting spheres.

Figure 3a is the correlation of two perfectly conducting, $r = 0.50$ μm spheres, illuminated at $\lambda = 647.1$ nm, as a function of separation distance d. We calculated the scattering intensities used in the correlations in the forward ($\theta = 0°$) and backward ($\theta = 180°$) scatter directions by averaging over the orientations of the two spheres with respect to the incident beam for each separation distance. Figure 3a indicates that the correlation is generally positive and tends toward zero as the separation distance increases. Also, oscillations in the correlation are similar to oscillations that would be seen in the scattered field due to interference; in fact, the frequency of these correlation oscillations is approximately equal to half the wavelength of the incident field.

We argue that these correlations are predominantly positive because of the geometry of the system. For small separations, scattering cross sections are generally greater when light is incident on two spheres in a broadside orientation than when the spheres are in an end-on incidence. This is due to electromagnetic flux intercepting the scattering object. A greater cross section tends to increase scattering intensities in all directions; hence, as the orientation changes from broadside to end-on incidence, the scattering intensities in both forward and backward directions tend to decrease, and the resulting correlations measured will be positive.

(a)

(b)
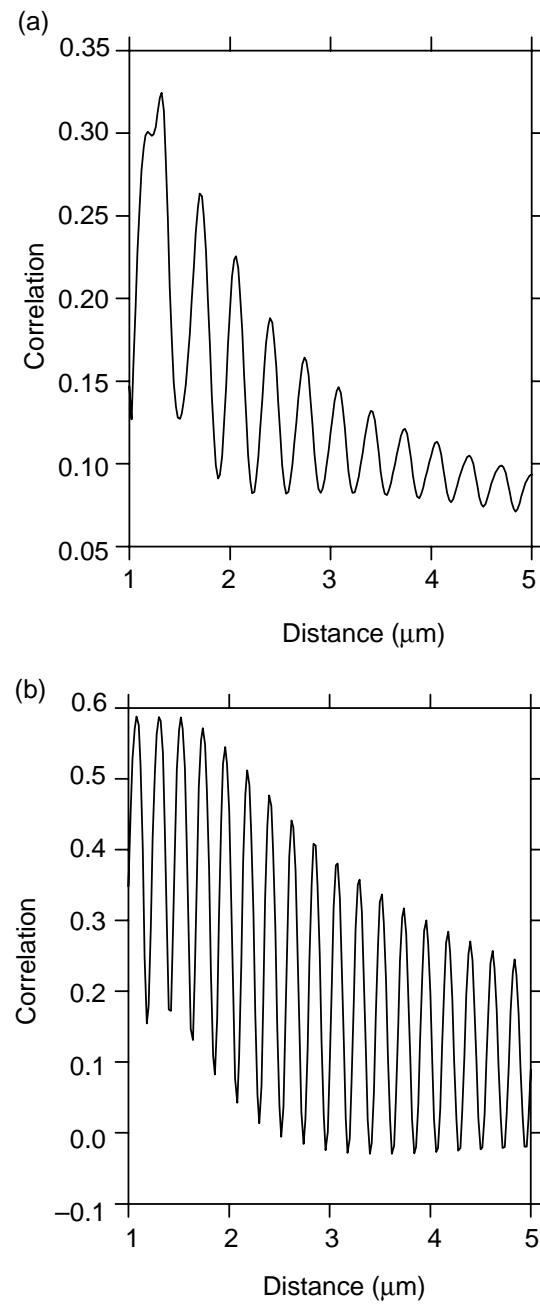
Figure 3. Correlation of (a) two perfectly conducting, $r = 0.50$ μm spheres illuminated at $\lambda = 647.1$ nm as a function of separation distance d, and (b) two dielectric, $n = n_i/n_h = 1.081$, $r = 0.50$ μm spheres illuminated at $\lambda = 439.9$ nm as a function of separation distance d.

Although these results provide some insight into the correlation, it is more appropriate to examine the correlation for the system parameters within the host sphere. In this case, the incident wavelength and refractive indices of the spheres are modified by the refractive index of the incident medium. Figure 3b shows the correlation of the light scattering of the two spheres ($n = n_i/n_h = 1.081$, $r = 0.50$ μm, illuminated at $\lambda = 439.9$ nm) as a function of separation distance d. The results are similar to those in figure 3a, except that the correlation and the frequency of oscillation are greater. Again, the frequency of oscillation is approximately equal to half the wavelength of the incident light. Several characteristics apparent in figure 3 also appear in other cases examined in this work: (1) the correlation function oscillates as sphere separation increases, and the amplitude of oscillation decays as the separation distance increases; (2) the frequency of oscillation is equal to approximately half the wavelength of the incident radiation; and (3) as the size of the spheres increases, the correlation tends to be more positive. Since the correlations for this model have predominantly positive values, we suggest that the experimental correlations observed (which display both positive and negative correlation) are not due to interference of waves scattered by inclusions.

The second type of system we examine is a host sphere containing a single nonconcentrically located spherical inclusion [40–48]. It has been hypothesized that the signal oscillations in our experiment are due to inclusions passing through regions of high and low intensity within the host sphere [13]. To explore this hypothesis, we examine the scatter from a host sphere containing a nonconcentrically located spherical inclusion. The correlation of backward and forward scatter is then due solely to the changing position of the inclusion within the host (orientation averaging is carried out for each position). Figure 4 shows the correlation of forward ($\theta = 0°$) and backward ($\theta = 180°$) scatter intensities, as a function of the center-to-center distance between a latex inclusion ($r_i = 0.50$ μm) and its glycerol host ($r_h = 2.000$ and 2.005 μm). Some characteristics apparent in these graphs appear in other correlations that we have examined from this type of system. In general, the correlation is dependent on the position of the inclusion within the host and is much more sensitive to changes in the size of the host when the inclusion is near the edge of the host. Unlike the correlations due to interference, the correlations for a droplet containing an inclusion have strongly negative as well as positive correlations.

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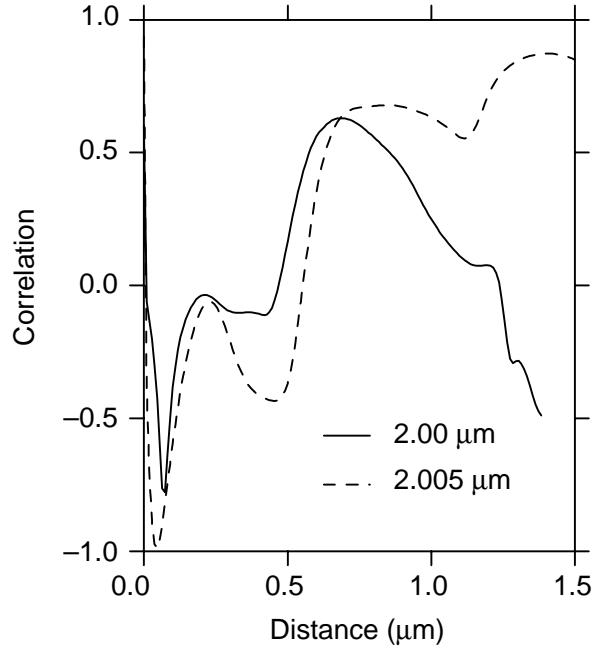
Figure 4. Correlation of glycerol host spheres ($n_h = 1.471$) containing an $r = 0.50$ μm latex sphere illuminated at $\lambda = 647.1$ nm as a function of center-to-center distance d.

One quantity that may be of interest is a spatial constant associated with the correlations. We can find this value by calculating the autocorrelation of the calculated cross-correlation function. The decay associated with the autocorrelation corresponds to a spatial constant over which the cross-correlation changes. These calculations were performed on data sets for different inclusion and host sphere radii. When the inclusion size remains smaller than the wavelength, these spatial constants depend primarily on the wavelength interior to the host sphere and have values on the order of $\lambda/4n_h$. These spatial constants are approximately the same as spatial constants that would be achieved from correlations of the two-sphere system. They do not, however, show the obvious periodicity seen in the two-sphere correlations.

Our analysis of these two relatively simple scattering models suggests that correlations due to inclusions diffusing through the host sphere resemble the experimental data sets that we have collected more closely than those of the isolated two-sphere model: in particular, the observed correlations vary over a wide range of both positive and negative values. This assertion is based on two assumptions: (1) that correlations calculated from a host sphere containing one inclusion do not differ substantially for a host sphere containing

multiple inclusions; and (2) that correlations due to interference among multiple spheres tend to remain positive, like those due to interference between two spheres.

Although we took steps to reduce droplet evaporation, we find it necessary to assess the effect of this small but nonzero evaporation. In order to do this, we examine model correlation calculations of a glycerol host containing a latex inclusion positioned at its outer edge (fig. 5a). We choose this particular configuration, since, as was seen in figure 4, it maximizes changes in the correlation function, and therefore puts an upper limit on the effect of droplet evaporation. Importantly, the correlation does not change appreciably, except for fairly narrow regions where the inclusion is positioned near the surface of the host, where it veers strongly toward +1. These regions of high positive correlation are due to resonances (shown in the extinction curve of fig. 5b). On resonance, scatter in virtually all directions is enhanced, so the light scattered in different directions has strong positive correlation. Thus, to avoid significant effects of even slow droplet evaporation on scattering, we need to avoid droplet resonances during our observations. We have taken this precaution in the data reported here.
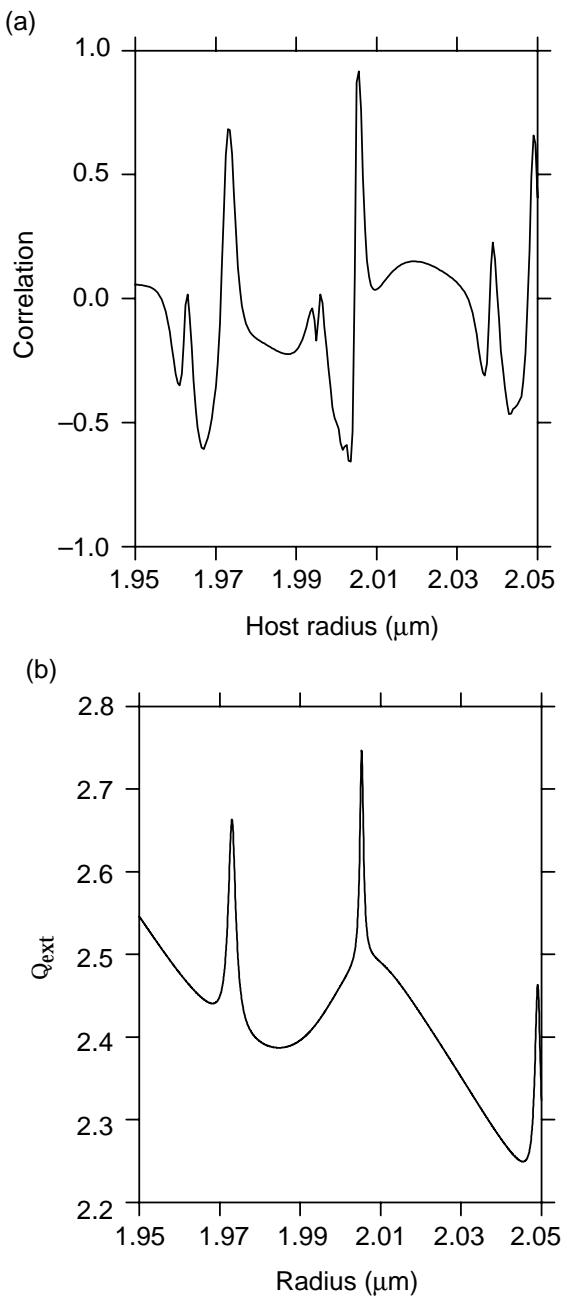
Figure 5. For a glycerol host sphere containing an $r = 0.50$ μm latex sphere fixed to its outer edge, (a) correlation as a function of host sphere radius, and (b) extinction as a function of homogeneous glycerol radius.

4. Conclusion

In this report, we describe a new method of analyzing the intensities from a scattering system. We have made experimental measurements of angular correlations and numerically calculated correlations from two different types of scattering systems. The numerical calculations suggest that we can distinguish between a scattering system composed of a conglomeration of spherical particles and one in which the system is enclosed by a spherical host. We observe that in a single levitated droplet containing inclusions, the fluctuations in the near-forward and near-backward scattering directions, over periods of about 1 s, can be strongly correlated, uncorrelated, or strongly anticorrelated. These results are consistent with the random Brownian motions of the inclusions in high and low intensities within the host sphere, rather than from interference effects between the inclusions. This suggests that the photon-correlation analyses, which are based on interference, would not produce valid results for this particular scattering system.

For the most part, the analysis presented is qualitative. We outlined the methodology of angular correlation and presented the correlation characteristics of some specific scattering systems. From these characteristics, we can determine some general information about the scattering system. One goal of light scattering studies is not only to determine qualitative system characteristics, but also to determine quantitative characteristics from the signals. We are currently pursuing research in this field. Preliminary results lead us to believe that inclusion size and concentration can be determined from the time constants associated with the angular correlations.

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The scattering phase functions of micrometer-size glycerol droplets containing spherical latex inclusions undergo random fluctuations with time. We measure scattering intensities in the near-forward and near-backward scattering directions and find them to have strong positive correlations during some time periods and strong negative correlations during other time periods. The characteristic time constants of these correlations are on the order of seconds. We calculate scattering correlations from two types of scattering systems. Correlations from an aggregate of two isolated spheres are generally positive, whereas correlations from a sphere containing a single spherical inclusion may be both positive and negative. Calculations of correlations from our experimental data are consistent with diffusion of inclusions within the host droplet, rather than interference effects between the inclusions.

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